Report #1

First Semi-annual Report of the American University

Solid State Group

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This is the first report of the American University Solid State Group. During the past six months we have been organizing and assembling. Two new graduate students, Mr. Choudhury and Mr. Pochobradsky, have joined us. Mr. Kalva, whose work at American University will be considered part of his PhD dissertation at Charles University in Prague, Czechoslovakia, will be with us for only one year, as will Professor Daniel, who holds an NSF Senior Foreign Scientist Visiting Professorship. Professor Cohen has been assembling equipment for the measurement of transport properties of magnetic semiconductors.

There has been some progress in physics; three theoretical projects were completed and submitted for publication, and the Mössbauer group has completed a study of $Pb_x Sn_{1-x} Se$, and has almost completed studies of a series of palladium and Heusler alloys. These will be discussed in turn.

Magnetoelastic Spin Hamiltonians: Application to Garnets

American University Investigator: Earl Callen

Others: Richard Alben, Yale University

The commonly accepted theories of the temperature and field dependence of magnetostriction[1] ignore crystal field effects. Yet in many materials these effects are far from negligible. And in some, in spite of the large crystal fields the experimental results seem to agree with the simple isotropic theory. A bewildering example is dyspropium iron garnet, in which one magnetostriction coefficient, λ_{111}, seems to agree with the "single-ion, zero crystal field" theory, while another, λ_{100}, clearly does not. The intent and achievement of our effort[2] was to develop a general theory of magnetostriction including crystal field effects, and to rationalize the diverse observations. We display the general form of magnetoelastic spin Hamiltonian for all the 32 crystal point groups. And we show that, depending upon the relative sizes of the several coefficients which occur in the theory, it is quite possible in the same crystal for some magnetostriction coefficients to be "anomalous", while other coefficients appear to be quite unaffected by the large crystal fields.

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1. Magnetostriction, Earl Callen, J. Appl. Phys. 39, 519 (1968)
2. Magnetoelastic Spin Hamiltonians: Application to Garnets, Richard Alben and Earl Callen, Phys. Rev. (to appear)

Oscillatory Magnetostriction in Multivalley Semiconductors, with Application to PbTe

American University Investigator: Earl Callen

Others: Henry Belson, J. R. Burke, U. S. Naval Ordnance Laboratory

In a large magnetic field the energy levels of conduction electrons condense onto well spaced Landau levels. As the magnetic field strength is changed, these levels successively pass through the Fermi surface, and all those electronic properties of the solid which depend upon the density of states at the Fermi level display de Haas-van Alphen $\frac{1}{H}$ oscillations. We have studied these oscillations in the magnetostriction of PbTe, to see what information they can provide as to the band structure of this useful material. We show[1] that the observed period of the oscillations is in agreement with the known extremal cross-section of the Fermi surface. The amplitudes of the oscillations tell us much more. The principle but not the exclusive contribution to the amplitude comes from intervalley charge transfer; Landau levels of one valley pass through the Fermi surface at a different field strength from those of a differently oriented valley. The crystal lowers its free energy by straining, the distortion and consequent deformation potential raising the Landau levels of one valley and transferring electrons to a lower unpopulated level of another. A second interesting effect is best described in terms of what we call "strain masses", measures of the distortions of the shape of the valleys per unit strain. As the magnetic field is changed, not only do the valleys move up and down in energy, they wiggle back and forth and grow long and short to minimize the free energy. From comparison with experiment we are able to evaluate the strain masses that describe these distortions.

An interesting sidelight to this investigation is the question of "carrier freezout" in PbTe. Several years ago, when know-how of the growth of good PbTe single crystals was not yet developed, Weinberg and Callaway[2] attributed an observed exponential temperature dependence of the nmr Knight shift to carrier freezout. Their work confirmed other observations.[3] Yet, recent measurements show no such effect. One explanation may be the poor quality of early materials, another is that somehow the Knight shift, which measures the net electron spin density at the nucleus, is seeing something different from the magnetostriction oscillations, deHaas-van Alphen or Shubnikov-deHaas oscillations, which sense all the electrons on the Fermi surface. To test this possibility we intend to observe any temperature dependence of the isomer shift in the Mössbauer spectrum, as this shift measures something like the Knight shift (i.e., the total electron charge density at the nucleus).

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1. Oscillatory Magnetostriction in Multivalley Semiconductors, with Application to PbTe, H. S. Belson, J. R. Burke, and E. Callen, Phys. Rev. (to appear)
2. I. Weinberg and J. Callaway, Nuovo Cimento 24, 190 (1962)
3. M. Matyas, Czech. J. Phys. 8, 301 (1958)

Néel Ferrimagnets in Large Magnetic Fields

American University Investigator: Earl Callen

Others: A. E. Clark, U. S. Naval Ordnance Laboratory

The rare earth ions in a rare earth ion garnet are coupled antiparallel to the iron system by an exchange field, H_{ex}, which is weak ($\sim$100 koe) compared to that of the Fe - Fe exchange ($\sim 10^4$koe). Suppose the iron moment $\vec{M}_{Fe}$ to dominate at 0^oK. Then in a small external field, $\vec{M}_{Fe}$ will point parallel to the field, $\vec{M}_R$ antiparallel. As the field is increased to infinity, all spins will ultimately lie along the field. There are two ways that $\vec{M}_R$ can get there, either by coherent rotation of the entire rare earth sublattice, or by paramagnetic dissolution and reformation of the rare earth moment in the net field $H-H_{ex}$. Both can happen[1]. This produces some weird moment vs. temperature and moment vs. field curves[1]. It also produces adiabatic magnetization cooling[2]; as the field is increased, the moment of the rare earth is decreased, the entropy increases and the crystal cools.

So far, we have investigated these effects only by molecular field theory, which is reasonably justified in the rare earth garnets, but not in ferrimagnets with more comparable exchange constants. We hope to perform a spin wave analysis of the H-T spin flop phase diagram for these other materials.

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1. A. E. Clark and E. Callen, J. Appl. Phys. **39**, 5972 (1968)

2. A. E. Clark and E. Callen, Phys. Rev. Letters **23**, 307 (1969)

Relation of Crystal Groups to the Macroscopic Properties of Solids

American University Investigators: Earl Callen and Z. Kalva

Others: H. B. Callen, University of Pennsylvania

The use of crystal character tables in analyzing the macroscopic properties of solids is so well established that it is surprising that until last year no one had given these tables in a form which made them immediately useable without a good deal of algebraic manipulation. While such tables in the simplest and most useful form have now been given[1], along with an admirably lucid explanation that makes use of the tables painless, even by those who know no group theory, the tables themselves are not entirely complete, and in some instances may be misleading. In collaboration with Professor H. B. Callen, we are revising and extending the tables and elaborating on the explanation of their application.

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1. H. B. Callen, Am. J. Phys. 36, 735 (1968)

Collective Electron Theory of the Metal-Semiconductor Transition in Magnetite

American University Investigator: E. Callen

Others: J. R. Cullen, U. S. Naval Ordnance Laboratory

Below the Verwey temperature ($T_V = 119^oK$), the Fe^{2+} and Fe^{3+} ions on the B sites in magnetite order on alternate (100) planes. At the transition the ions disorder, and the conductivity increases by a factor of 100. We give[1] a band theoretical explanation of this phenomenon. Since it is the electrons themselves that make the difference between an Fe^{3+} and Fe^{2+} ion, one can say that in the ordered states the electrons themselves break the lattice symmetry by doubling the unit cells, piling up on every other site, creating a new Coulomb periodic potential in which they self-consistently order. They do this to lower their correlation energy. We show that this reduction of the Brillouin zone exactly fills the central zone and the lowest energy band, and produces an insulator. The theory is BCS-like, with an order parameter (number of electrons on even sites minus number on odd sites) and gap which should show a second order phase transition, and an exponential dependence on density of states and coupling constant in the weak coupling limit. We intend to study the electrical conductivity, thermoelectric power, and deviations from stoichiometry.

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1. J. R. Cullen and E. Callen, J. Appl. Phys. (to appear)

Phase Change in SnTe

American University Investigators: R. Segnan and W. A. Ferrando

Others: B. Houston, H. Savage, U. S. Naval Ordnance Laboratory

For several years there has been a controversy whether there is a crystal structure change in SnTe at low temperatures. We hope we can resolve this by observation of the temperature dependence of the Mössbauer isomer shift. There is ordinarily a linear dependence of this shift on temperature, due to thermal expansion. A phase change, modifying the crystalline environment, the wave functions, and hence the charge density at the nucleus, may cause a departure from the background linear relationship. Experiments are in progress.

Band Crossing in $Pb_x Sn_{1-x} Se$

American University Investigators: R. Segnan and W. A. Ferrando

Others: J. Dixon, G. Hoff, U. S. Naval Ordnance Laboratory

As one varies either the compositions or the temperatures of $Pb_x Sn_{1-x} Se$, the conduction and valence bands at the L point of the Brillouin zone, bands which behave differently under inversion, gradually reverse in energy. Thus, if the positive band is higher in energy on one side of the transition, then the gap narrows when the temperature is changed, is zero at the band inversion temperature, and the negative band is higher beyond this temperature. As one traverses the band inversion temperature, the transport properties, due to electrons on the Fermi surface, alter. We wondered if the isomer shift in the Mössbauer absorption would also betray the band inversion. It does not. Presumably no measurable change in the number of electrons with S-like character accompanies an inversion of the bands.

Mössbauer Studies of Ni-Pd Alloys

American University Investigators: R. Segnan and W. A. Ferrando

Others: A. I. Schindler, U. S. Naval Research Laboratory

The role of magnetic impurities (Fe, Co) in pure palladium or in palladium alloys is being investigated by means of Mössbauer measurements. In most cases, bulk magnetization measurements leave uncertainties as to the impurity and matrix contributions to the polarization effects. In order to determine the impurity contribution to the polarization, we examine the effect of small amounts (0.01% or less) of iron and cobalt in a series of Ni-Pd alloys. From the same data, we were able to determine polarization effects of the nickel palladium matrix at various concentrations.

By studying the hyperfine fields at ^{57}Fe nuclei, we find evidence of impurity clustering polarization which we can separate from the Ni-Pd matrix polarization. The impurity polarization effects are reduced by long term anneal of the samples.

Mössbauer Effect Studies in Heusler Alloys

American University Investigators: E. Daniel, R. Segnan, and W. A. Ferrando

Mössbauer effect measurements were performed at the nuclei of Sn^{119} in the ferromagnetic Heusler alloys Cu_2MnSn and $Cu_{2.01}Mn_{0.89}Sn_{1.10}$. The samples, used as absorbers in powder form, were subjected to heat treatments as described in (1). Mössbauer spectra were taken after each phase of the heat treatment, i.e., (a) without annealing the sample, (b) by annealing the bulk sample, (c) by annealing the powdered sample. The measurements were performed at various temperatures ranging from about 57° K to 375° K. A hyperfine field of the order of 20 kOe [about 1/10 of that reported in (1)] was observed at 77° K. By increasing the temperature, the hyperfine field decreased to about 6 kOe at room temperature. The heat treatments indicated little variation in the measured hyperfine fields. Further measurements are being performed.

A rough estimate by E. Daniel, following the theory of Blandin and Caroli, gives for the hyperfine field at the ^{119}Sn nucleus, a value of the order of 100 kOe and a negative sign.

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(1) V. V. Chekin, L. E. Danilenko, and A. I. Kaplienko, Soviet Phy. JETP, 24, 472 (1967)